

Research on the rules of influence of transparent agent infiltration on adsorption and diffusion of coal petrography gas

The paper aims at studying the influence of rules of transparent agent infiltration of coal seam on coal seam gas diffusion. With coal particles in Gecun village, Henan Province as research object, it makes use of adsorption and desorption experiments in high pressure and variable temperature for isothermal and desorption experiments that is based on three common transparent agents, fenton reagent, acetic acid and hydrochloric acid, comparing gas of coal particles before and after infiltration. Moreover, the thesis is on the basis of the new diffusion model of dynamic diffusion coefficient and applies MATLAB software to programming procedure for calculating the original diffusion coefficient D_0 of the gas desorption experiment and its attenuation coefficient β . The experimental results are as follows: transparent agents can inhibit gas adsorption of coal particles, and the inhibitory effects of these transparent agents grow stronger from acetic acid, hydrochloric acid to fenton reagent; transparent agents can promote gas diffusion of coal particles, and the inhibitory effects of these transparent agents grow stronger from acetic acid, hydrochloric acid to fenton reagent; transparent agents in coal seam can mitigate outburst risks of coal seam gas with acetic acid the best effect.

Keywords: Adsorption, coal particle, desorption, infiltration, transparent agent.

Introduction

Outburst of coal petrography gas tops the six major disasters of coal mine and is a challenging obstacle to secure and effective production of coal mine in China [1]. Gas extraction is one of the major actions for controlling outburst of coal petrography gas. Nevertheless,

low permeability, high gas pressure and strong gas adsorption of coal seam in China hamper efficiency of gas extraction and bring modest extraction result. Some transparency actions like hydraulic fracturing, hydraulic punching, deep hole blasting, and phase change fracturing of carbon dioxide are solutions to low permeability of coal seam. Guo Hongyu and Su Xianbo [2] during the research on peranthracite in Jiaozuo, Henan Province found out that non-completely closed endogenous fractures of coal and even part of exogenetic fractures are mostly filled with calcite, thus come up with that acidification infiltration method is superior to fracturing in theory.

Guo Hongyu and Su Xianbo [2] via microscope observe that coal samples with various metamorphic grades soak in chlorine dioxide can be etched differently with the low rank coal the most sever etch. Furthermore, the comparison test of permeability explores that chlorine dioxide can improve permeability in coal reservoir, and the better permeability an original coal reservoir has, more is prominent increase of permeability after soaking in chlorine dioxide. Beall [3] applied two different acid fluid systems to infiltrating coal petrography as well as compared and test permeability and porosity of coal core before and after infiltration. Chinese scholars including Guo Hongyu and Su Xianbo [2], Li Rui [4], Zhao Wenxiu [5] and Ni Xiaoming [6] utilized various acid fluids to conduct acidification infiltration for coal reservoir, and found out that acidification infiltration dissolves and corrodes clay mineral of fractures in coal reservoir as well as changes pore structure and surface properties of coal seam.

The previous scholars mainly focus on the influence of common acid transparent agents like fenton reagent, acetic acid and hydrochloric acid on infiltrating features of coal seam [7], but seldom mention desorption-diffusion rules of effect of transparent agent on coal petrography gas. Coal seam gas cannot be extracted without three phrases, namely desorption, diffusion and transfusion [8-13]; therefore, it is of vital importance to study the effect of transparent agent on the desorption and diffusion rules of coal petrography gas. The thesis based on previous research aims at studying desorption influence of transparent agent infiltration on gas

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of coal particles. With coal particles in Jiulishan mine, Henan Province as research object, this paper applies adsorption and desorption experiments in high pressure and variable temperature for isothermal and desorption experiments involving three common transparent agents, fenton reagent, acetic acid and hydrochloric acid, as well as comparing gas of coal particles before and after infiltration. In addition, it also makes use of new physical model of gas diffusion in multi-scale pore of coal particle to analyze the mechanism of desorption effect of transparent agent infiltration on gas of coal particles, the result of which can become theoretical support for transparent actions in coal seam.

2. Experiment research

2.1 EXPERIMENT SYSTEM AND SAMPLE PREPARATION

The isothermal and desorption experiment of gas of coal particles is based on the adsorption and desorption experiment system in high pressure and variable temperature which is an independent research fruit of Henan Polytechnic University (HPU). The equipment is mainly composed of adsorption system, desorption system, temperature-pressure measurement and control system, as well as computer data collecting system. The schematic diagram of the device is illustrated in Fig.1.

On the basis of research demand, the experiment takes coal at Gecun village in Yima coal mine as the samples that are selected with diameter of 1-3mm and are ready for use after drying for 4 hours. The device for soaking coal samples is MesoMR23-060H-I vacuum device made by Key Laboratory of Gas Geology and Gas Control in HPU. The infiltration preparations are fenton reagent, acetic acid, hydrochloric acid and pure water (as control group).

2.2 EXPERIMENTAL PROCEDURES

1. Wash and filter coal particles having been through acid

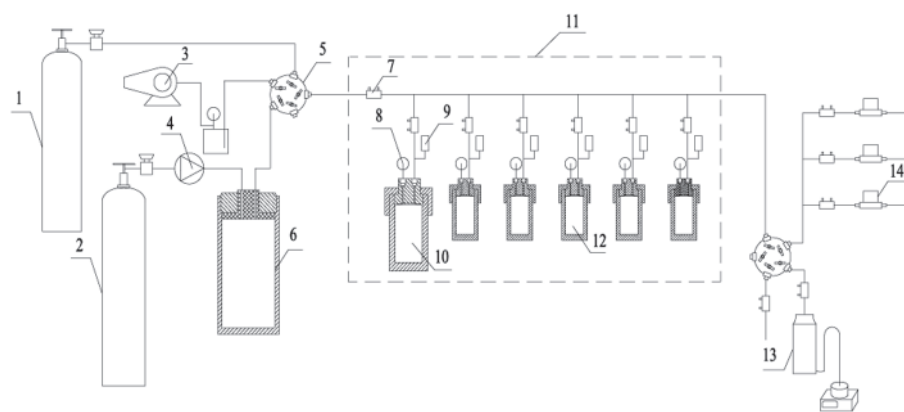


Fig.1 Schematic diagram of adsorption and desorption device in high pressure and variable temperature

1-Helium gas cylinders, 2-Methane gas cylinders, 3-Vacuum pump, 4-Booster pump, 5-Six valve, 6-The methane tank, 7-Pneumatic valve, 8-Thermometer, 9-Pressure gauge, 10-Gas buffer tank, 11-Calorstat, 12-Coal sample tank, 13-The method of drainage gas metering device, 14-Gas meter

infiltration, dry the coal particles in thermostatic drying oven for 48 hours, and put coal samples into each sample jar. To avoid coal samples block pipeline during vacuum degassing, while recording quality of each coal sample, experimenters should lay a layer of pledget and 80 mesh of cooper grid on coal sample.

2. Open vacuum pump to conduct vacuum drainage for each coal sample until vacuum degree reaches 20 Pa and lasts for 2 hours before closing the pump and finishing degas.
3. Test the constants of coal particles after infiltration according to ref. [14].
4. Adjust experimental temperature to $(30 \pm 1)^\circ\text{C}$, and use methane gas cylinders and buffer gas to aerate coal sample respectively. The pressure after adsorption equilibrium of coal sample in this experiment is 1.1 MPa.
5. Use mass-flow gas meter to measure gas desorption volume and gas desorption rate in different adsorption equilibrium pressures of each coal sample, and compare and analyze the influence of desorption rules of acid infiltration on gas of coal particles.

3. Experiment results

3.1 INFLUENCE OF TRANSPARENT AGENT INFILTRATION ON GAS ADSORPTION

After infiltrating coal particles respectively into clear water, fenton reagent, hydrochloric acid and acetic acid, results of gas adsorption were carried out with Langmuir equation. Fig.1 shows gas adsorption parameters of the four samples.

Table 1 demonstrates different maximum gas adsorption capacities of infiltrated coal particles. For the samples infiltrated in clear water, the maximum capacity is 19.705ml/g; acetic acid, the maximum capacity is 12.987 ml/g, 34.1% decrease of that in clear water; hydrochloric acid, 13.724 ml/g,

30.3% decrease of that in clear water. These figures show that transparent agents can inhibit gas adsorption of coal particles, and the inhibitory effects of these transparent agents grow stronger from fenton reagent, hydrochloric acid to acetic acid.

3.2 INFLUENCE OF TRANSPARENT AGENT INFILTRATION ON GAS DESORPTION

After infiltrating coal particles into various transparent agents, variation trends of gas desorption are demonstrated in Fig.2.

Fig.2 shows though at different stages, similar tendency of gas desorption can be detected in different samples, that is, a fast growth in the beginning, a continuous increase in

TABLE 1: GAS DESORPTION PARAMETERS OF COAL PARTICLES INFILTRATED INTO TRANSPARENT AGENTS

Infiltration reagent	Adsorption constant a (ml/g)	Adsorption constant b (MPa ⁻¹)
Clear water	19.705	0.675
Fenton reagent	16.482	0.503
Acetic acid	12.987	0.624
Hydrochloric acid	13.724	0.509

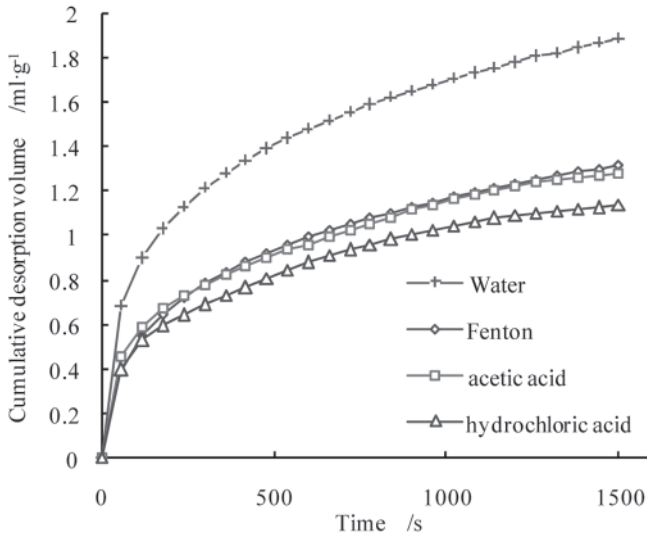


Fig.2 Gas desorption curves of coal particles infiltrated into transparent agents

the middle and a steady movement at the end. Total desorption amount differs in the same stage. Among all the samples, the coal particle infiltrated in clear water takes the maximum amount, while the samples in hydrochloric acid the minimum amount. Fenton reagent and acetic acid show the similar diffusion amount. When the adsorption equilibrium pressure is 1.1 MPa, maximum gas diffusion amount of the samples are 8.13 ml/g, 7.29 ml/g, 4.62 ml/g, 4.37 ml/g, 4.07 ml/g calculated with gas diffusion parameters of coal particles infiltrated into transparent agents. Fig.3 shows time tendency of calculable gas diffusion ratio of coal particles infiltrated into transparent agents.

Fig.3 shows that the effects of these transparent agents become weaker from acetic acid, fenton reagent, indicating transparent agents can accelerate gas desorption of coal particles, and hydrochloric acid to clear water, and coal particles infiltrated into acetic acid holds the largest amount of gas desorption.

4. Analysis and discussion

To conduct a quantitative analysis on the influence of transparent agent infiltration on gas desorption, this chapter, based on dynamic diffusion coefficient model put forward by Li Zhiqiang, analyzes the initial diffusion coefficient D_0 and attenuation coefficient β of gas contained in coal particles

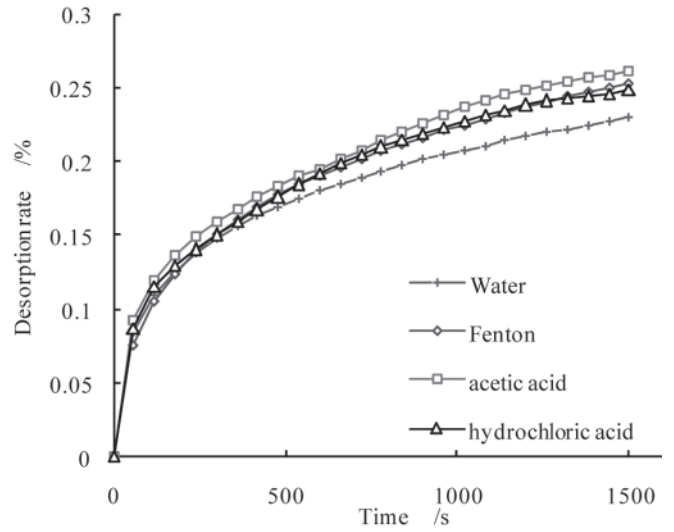


Fig.3 Gas desorption curves of coal particles infiltrated into transparent agents

infiltrated into transparent agents, to study the disciplinary influence of transparent agents infiltration on gas diffusion-filtration in coal particles.

4.1 NEW MODEL ON DYNAMIC DIFFUSION

A large number of scholars from home and abroad agree on Fick Diffusion Law in gas diffusion. In the process, diffusion resistance remains unchanged as a constant. However in 1970s, researchers found that Fick Diffusion Law could only describe the early 10 minutes in diffusion process of tectonically undeformed coals. Therefore, recently Li Zhiqiang put forward a new model on dynamic diffusion which can accurately describe the whole gas diffusion process of all categories of coals under different conditions. This is the formula [15-16]:

$$\frac{Q}{Q_\infty} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left(\frac{n^2 \pi^2 D_0}{R^2 \beta}\right) (1 - \exp(-\beta t)) \quad \dots \quad (1)$$

In this formula, Q_t , cm³/g, is the accumulated amount of gas diffusion at the moment t ; Q_∞ , cm³/g, is gas desorption capacity under normal conditions; D_0 , cm²/s, stands for the initial diffusion coefficient when $t = 0$; R is the radius of coal particles, and in this paper, $R = 0.17$ cm; attenuation coefficient β , s⁻¹; t is the time, and s refers to non-quantity diffusion time T_D .

$$T_D = \frac{D_0}{R^2 \beta} (1 - \exp(-\beta t)) \quad \dots \quad (2)$$

Substituting the equation (2) into the equation (1) gives:

$$\frac{Q}{Q_\infty} = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} e^{-n^2 \pi^2 T_D} \quad \dots \quad (3)$$

The equation (3) shows that according to variation of gas desorption of coal particles, factor theorem can be used to work out the diffusion law of non-quantity diffusion time T_D . Based on equation (2), the way of nonlinear curve fitting to analyzing non-quantity diffusion time T_D can be used in calculating initial diffusion coefficient D_0 and attenuation coefficient β .

Factor theorem can be used to work out the diffusion law of non-quantity diffusion time T_{Di} at the moment of t_i , which is demonstrated in Fig.4.

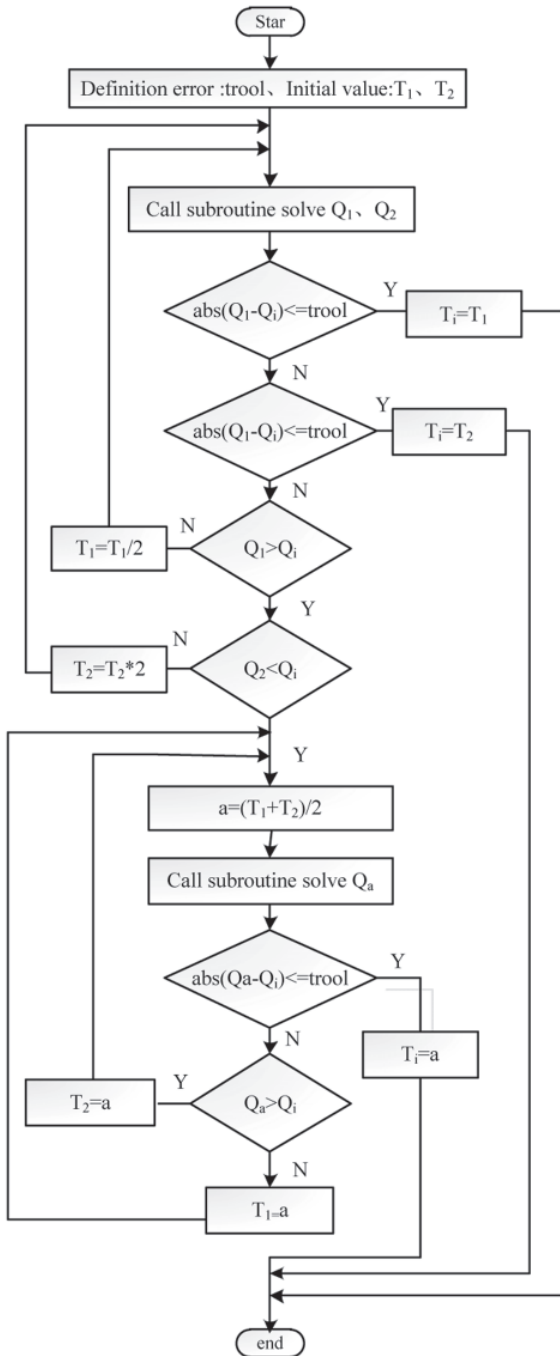


Fig.4 Flow diagram of calculation of non-quantity diffusion time

4.2 GAS DIFFUSION FEATURES OF COAL PARTICLES INFILTRATED IN TRANSPARENT AGENTS

Gas desorption of coal particles infiltrated in transparent agents share the same laws among different transparent agents, so this paper takes coal particles in fenton agent as an example to demonstrate the calculation of initial diffusion coefficient D_0 and attenuation coefficient β .

Factor theorem can be used to work out the diffusion law of non-quantity diffusion time T_D at the moment of t and exponential functions fitting to analyzing the variation curve of T_D at different time, which is demonstrated in Fig.5.

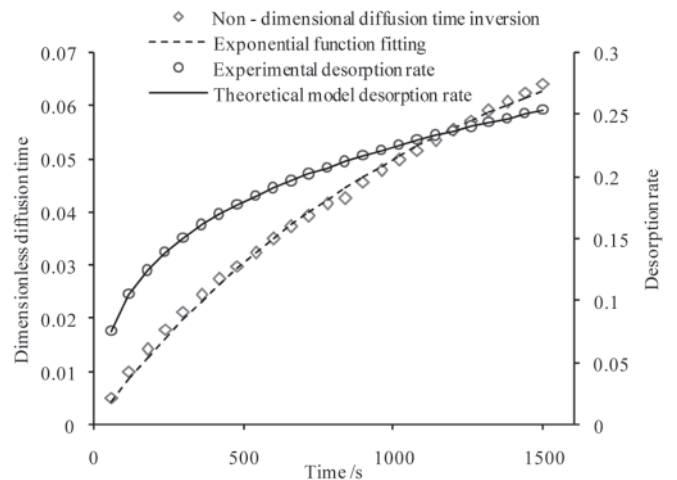


Fig.5 The variation curve of non-quantity diffusion time T_D at different time

According to Fig.5, the experiment result is highly identical with gas desorption rate under the model of dynamic diffusion model, with the correlation coefficient being 0.9998. Exponential functions fitting to analyzing initial diffusion coefficient D_0 work out the results as $2.17 \times 10^{-6} \text{ m}^2/\text{s}$ and attenuation coefficient β , $8.79 \times 10^{-6} \text{ s}^{-1}$. By the same token, initial diffusion coefficient D_0 and attenuation coefficient β of gas desorption of coal particles infiltrated in transparent agents are shown in Table 2.

References [15-16] explain that dynamic diffusion model, which shows the decrease of gas diffusion parameters of coal particles, can describe multi-scale porosity diffusion model. And the references have also put forward the attenuation

TABLE 2: DIFFUSION PARAMETERS OF GAS OF COAL PARTICLES INFILTRATED IN TRANSPARENT AGENTS

Infiltration reagent	The initial diffusion coefficient D_0 (cm^2/s)	Attenuation coefficient B (s^{-1})
Clear water	2.18E-06	1.29E-03
Fenton reagent	2.17E-06	8.79E-04
Acetic acid	2.39E-06	8.91E-04
Hydrochloric acid	2.33E-06	1.02E-03

mechanism of diffusion parameters of multistage series porosity. Table 2 shows that after being infiltrated into hydrochloric acid and acetic acid, coal particles shows a certain increase in the initial gas diffusion coefficients and a decrease in attenuation coefficients, which prov the function of transparent agents in reaming coal petrography. This results agrees with the precedent research that transparent agents can promote infiltration of coal seam. The gas diffusion parameters in Table 2 is available in calculating the variation of the efficient gas diffusion parameters of coal particles infiltrated in transparent agents, and the result is shown in Fig.6.

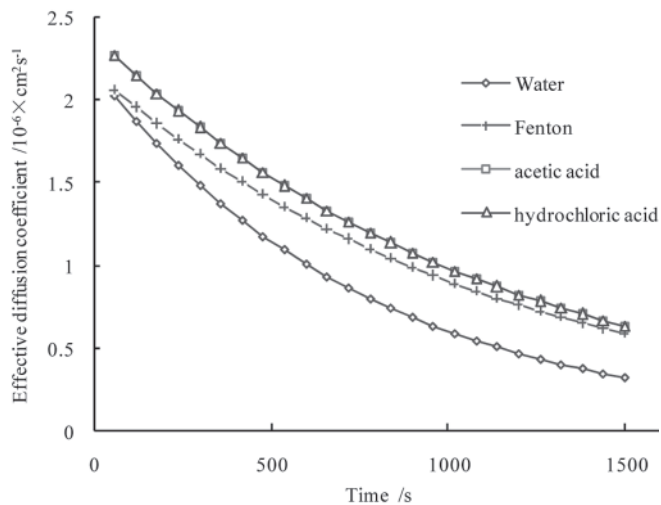


Fig.6 Variation curve of the efficient gas diffusion parameters at different time

Fig.6 demonstrates that after being infiltrated into hydrochloric acid and acetic acid, coal particles shows a certain increase in the efficient gas diffusion coefficients. It proves that transparent agents infiltration can promote gas diffusion and hydrochloric acid and acetic acid are the most effective agents.

Above all, transparent agents can inhibit gas adsorption and promote gas diffusion, so the infusion of transparent agents can decrease gas adsorption and increase gas permeability, facilitate gas extraction and then much mitigate the risk of coal and gas outburst. From the analysis above, conclusion can be drawn that compared with hydrochloric acid and fenton agent, acetic acid is more efficient in inhibiting gas adsorption and promoting gas diffusion. Therefore, concerning the influence of gas adsorption and desorption, acetic acid is proved to be a better choice in gas controlling.

5. Conclusion

1. Transparent agents can inhibit gas adsorption of coal particles, and the inhibitory effects of these transparent agents grow stronger from acetic acid, hydrochloric acid to fenton reagent.

2. Transparent agents can promote gas diffusion of coal particles, and the inhibitory effects of these transparent agents grow stronger from acetic acid, hydrochloric acid to Feton reagent.
3. Acetic acid is proved to be a better choice in gas controlling.

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